

Spontaneous chemical ordering and exchange bias in epitaxial $\text{Mn}_{0.52}\text{Pd}_{0.48}/\text{Fe}(001)$ bilayers prepared at room temperature

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We report spontaneous chemical ordering of $\text{Mn}_{0.52}\text{Pd}_{0.48}$ films, grown by molecular-beam epitaxy on body-centered-cubic $\text{Fe}(001)$ films. The bilayers were grown at room temperature onto a seeding structure of $\text{Ag}(001)/\text{Fe}(001)/\text{GaAs}(001)$. X-ray, neutron, and electron diffraction studies confirm extensive chemical ordering of the MnPd to the L1_0 antiferromagnetic phase which grows as a twinned film with the tetragonal c axis in the film plane. The Fe film exhibits a unidirectional exchange bias and we find no pronounced difference in magnetic structure of the biased Fe layer at the ascending and descending crossing fields where the net moment along the applied field axis is zero. © 2002 American Institute of Physics. [DOI: 10.1063/1.1436286]

The phenomenon of exchange biasing¹ in ferromagnetic/antiferromagnetic (FM/AF) bilayers is of intense current interest because of its widespread application in spin valves and magnetic tunnel junctions. Despite this activity, the physical basis of exchange biasing remains incompletely understood.² Exchange biasing has been studied³ in model epitaxial FM/AF material systems which incorporate low-temperature antiferromagnets such as FeF_2 and MnF_2 . However, in practical applications, antiferromagnets with Néel temperatures far above room temperature are required. The understanding of the phenomenon in these cases is far less developed.

A class of such antiferromagnets are the alloys MnNi , MnPt , and $\text{Mn}(\text{Pt,Pd})$. These are antiferromagnetic in the chemically ordered L1_0 (CuAu I) phase. Several issues remain unresolved, including the dependence of exchange bias on the crystallographic orientation of the AF in contact with the FM and dependence of exchange bias on the extent and length scale of chemical ordering. Fe/MnPd bilayers were introduced by Tang *et al.*^{4,5} as a model system for study since MnPd is a high (540 °C) Néel temperature L1_0 AF and the geometric match of its basal plane to body-centered-cubic (bcc) Fe ($a_{\text{MnPd}}/\sqrt{2}=2.878$ Å; $a_{\text{Fe}}=2.866$ Å) suggests that MnPd (001) can be grown epitaxially on bcc Fe with the c axis along the film normal (c -axis growth). However, we have shown⁶ that c -axis epitaxy and a -axis epitaxy [$\text{MnPd}(100)\|\text{Fe}(001)$] can be achieved at a growth temperature of 200 °C on bcc $\text{Fe}(001)$, the growth orientation depending on the surface morphology of the Fe film. Similarly, Cheng *et al.*⁷ reported a -axis and c -axis epitaxy of MnPd on bcc $\text{Fe}(001)\|\text{MgO}(001)$ grown by sputtering. In this latter work, the substrate temperature was held at 200 °C for a -axis growth and 300 °C for c -axis growth. At these elevated temperatures, significant interdiffusion is inevitable and the no-

tion of an abrupt FM/AF interface, with a well-defined chemical and magnetic structure, does not hold.

Here, we show that a -axis epitaxy of MnPd on bcc $\text{Fe}(001)$ can be achieved at room temperature. Spontaneous chemical ordering and exchange biasing are demonstrated. This result has technological significance since all L1_0 antiferromagnets currently used in applications require deposition or field annealing at temperatures above 200 °C to establish exchange bias.

The bilayers were grown by molecular-beam epitaxy (MBE) on $\text{GaAs}(001)$ wafers. A magnetic bias field of ~ 50 Oe along a $\text{GaAs}[100]$ direction was present during growth. A seed layer of ~ 20 Å Fe was deposited on the $\text{GaAs}(2\times 4)$ surface with the substrate at 180 °C, followed by a $\text{Ag}(001)$ film at 100 °C. The sample was cooled to 30 °C and a bilayer, consisting first of Fe then MnPd , was grown. Reflection high-energy electron diffraction (RHEED) patterns showed relatively sharp streaks confirming epitaxial growth $\text{Fe}(001)\|\text{Ag}(001)\|\text{Fe}(001)\|\text{GaAs}(001)$. However, the RHEED pattern from MnPd showed diffuse lines resulting from a superposition of two structural domains with the $\text{MnPd } c$ axis parallel to $\text{Fe}[110]$ or $[1\bar{1}0]$. A capping layer of Au was deposited on the MnPd . The MnPd composition was determined by Rutherford backscattering (RBS) to be $52 \pm 2\%$ Mn and the composition profile of the sample determined from a fit^{8,9} to the specular x-ray reflectivity data (see Table I). The interfaces between the layers were modeled as error functions with variable full widths at half maximum that ranged from 12 to 16 Å.

X-ray diffraction scans confirmed the $\text{MnPd } c$ axis was in-plane and twinned with $\text{MnPd}[001]\|\text{Fe}[110]$, $\text{Ag}[100]$ or $\text{Fe}[1\bar{1}0]$, $\text{Ag}[010]$. The average long-range chemical order parameter (S) was determined¹⁰ to be 0.85 ± 0.05 . Measured lattice constants of MnPd were $a(\text{normal})=4.05 \pm 0.01$, $a(\text{in plane})=4.054 \pm 0.006$ Å and $c(\text{in plane})=3.620 \pm 0.005$ Å. These are similar to bulk values¹¹ ($a=4.07$ Å

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TABLE I. Properties of epitaxial MnPd/Fe samples.

Sample No. Growth T	Film normal	H_E/H_C (Oe)	t_{Fe}/t_{MnPd} (Å)	Sample No. Growth T	σ (erg/cm ²) ± 0.005	S
1333 ^a 30 °C	a axis (1.0) ^c	$32 \pm 2/75 \pm 2$	$50 \pm 5/388 \pm 15$	1333 ^a 30 °C	0.027	0.85 ± 0.05
1355A ^b 200 °C	c axis (0.6 ^c)	$29 \pm 2/37 \pm 2$	$85 \pm 10/259 \pm 20$	1355A ^b 200 °C	0.042	0.9 ± 0.1
1355B ^b 200 °C	a axis (0.68) ^c	$46 \pm 2/86 \pm 5$	$85 \pm 10/259 \pm 20$	1355B ^b 200 °C	0.067	1.0 ± 0.1

^aProfile from x-ray reflectivity: Au(27 ± 2)/Mn_{0.52}Pd_{0.48}(388 ± 15)/Fe(50 ± 5)/Ag(295 ± 5)/Fe(20 ± 10)/GaAs(001).

^bProfile from RBS: Au(22 ± 2)/Mn_{0.52 \pm 0.02}Pd_{0.48 \pm 0.02}(259 ± 20)/Fe(85 ± 5)/MgO(001).

^cApproximate volume fraction of the MnPd film with the stated orientation.

and $c = 3.59$ Å) of Mn_{0.52}Pd_{0.48}. High-angle neutron diffraction measurements of the 388 Å-thick MnPd layer⁶ revealed a growth-axis (100) magnetic reflection and thus the MnPd layer is antiferromagnetic at room temperature. We suspect the film orders as a collinear antiferromagnet similar to bulk MnPd.¹²

Transmission electron diffraction patterns from plan-view samples confirmed twinning of the MnPd film and showed [Fig. 1(a)] strong (001) superstructure spots resulting from the superposition of (001) spots from the two structural domains. The absence of (110)-type spots suggests the absence of structural domains with the c axis perpendicular to the film plane. From both plan-view and cross section micrographs the average size of the in-plane domains of the MnPd was ~ 50 Å. High-resolution and diffraction contrast images of the FM/AF interface were consistent with a chemically sharp interface of width ~ 5 Å. (The x-ray reflectivity roughness is larger since it measures both physical and chemical intermixing and averages across a larger region.) Interfacial misfit dislocations were observed, suggesting relaxation of misfit strain.

Figure 1(b) shows longitudinal magneto-optical Kerr effect (MOKE) loops recorded with the applied field along the four in-plane Fe{100} symmetry directions. For the field applied along the bias direction ($\phi = 0^\circ$) the loop is shifted to negative field by 32 Oe. Reversal of this shift when the field was reversed ($\phi = 180^\circ$) confirmed the unidirectional nature of the bias. Along orthogonal easy directions of Fe ($\phi = 90^\circ$ and 270°) a double loop was observed. For these directions, the field axis is orthogonal to the bias direction. At sufficiently high fields, the magnetization saturates along the field direction. However, as the field is reduced, the magnetization rotates to the bias direction because this is the single equilibrium magnetic state of the Fe film in zero field. Finite coercivity causes this transition to occur at nonzero field. By using vibrating sample magnetometry, with two orthogonal sets of pick-up coils, we confirmed that at the crossing points (zero component of magnetization along the field axis) the Fe moment was oriented along the bias direction.

At room temperature, we also measured the field dependence of the neutron reflectivity for the a -axis MnPd sample for fields applied parallel, perpendicular and at 45° relative to the bias direction. For these measurements we used 4.75 Å neutrons on the NG-1 reflectometer at the NIST Center for Neutron Research and measured the nonspin-flip (NSF) reflectivities, R^{++} and R^{--} , and the spin-flip (SF) reflectivities, R^{+-} and R^{-+} using techniques detailed elsewhere.¹³ (The + and - signs in the superscript denote the polariza-

tion state of the incident and scattered neutron spins, which are aligned either parallel, +, or antiparallel, -, to the applied field.) The NSF reflectivities sense the chemical structure of the multilayer, and the splitting between R^{++} and R^{--} is sensitive to the component of the in-plane magnetization parallel to the applied field. The SF reflectivities provide information primarily about the projection of the in-plane magnetization perpendicular to the applied field. Figure 2 shows reflectivity data for a field of 29 Oe applied perpendicular to the bias direction (90° and 270°) after saturation. The data exhibit Kiessig fringes with spacing inversely proportional to the layer thicknesses. The SF scatter-

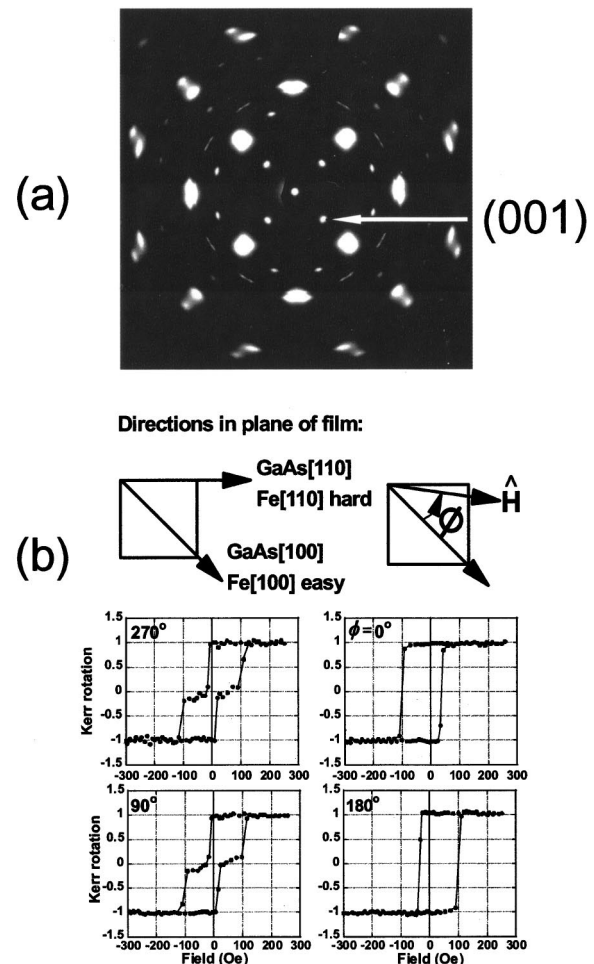


FIG. 1. (a) Transmission electron diffraction pattern from plan-view sample. Note four superstructure MnPd(001) spots which arise from MnPd twins, each having c -axis in-plane. (b) MOKE loops (longitudinal Kerr geometry, He-Ne laser) recorded for field along easy directions of Fe in plane. The (unidirectional) bias direction is defined as Fe[100], $\phi = 0$.

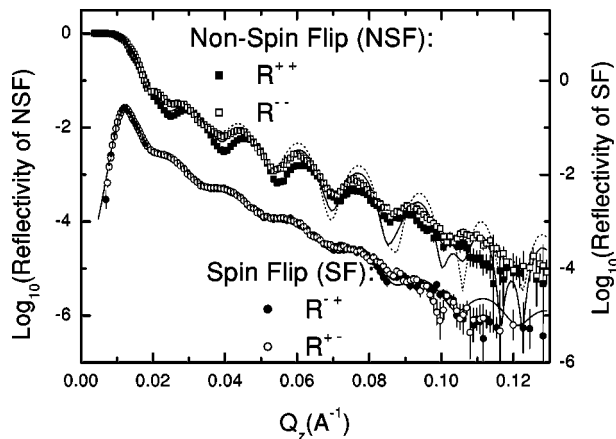


FIG. 2. Polarized neutron reflectivity as a function of the wave vector Q_z . Data were obtained in a field of 29 Oe applied perpendicular to the bias direction (90° and 270°) after saturation in a -400 Oe field. The data were corrected for instrumental background and the efficiencies of the polarizing elements. The fits are shown as solid and dashed lines.

ing is strong and the NSF reflectivities, R^{++} and R^{--} are nearly coincident. Fits to the data (solid lines) reveal that the Fe layer moment is fully aligned at 4° relative to the bias direction (nearly perpendicular to the applied field). We note that the Fe seed layer has a small moment oriented approximately 50° relative to the bias direction.

Magnetization and neutron reflectivity measurements both indicate that Fe switching occurs via moment rotation for fields applied perpendicular to the bias direction (90° and 270°). For fields applied parallel to the bias direction (0° and 180°), small splitting between the R^{++} and R^{--} reflectivities and the absence of SF scattering in neutron reflectivity data obtained at the crossing fields indicate that switching of the Fe moment direction proceeds via domain formation. In addition, the data did not show any pronounced differences in the magnetic characteristics of the Fe layer at the positive and negative field crossings. These results contrast with studies of epitaxial Fe/FeF₂ and Fe/MnF₂ films¹⁴ which show that the Fe switching at the negative crossing proceeds via moment rotation, while the switching at the positive crossing occurs via domain formation.

Our finding of an exchange bias for a -axis epitaxial MnPd/Fe(001) bilayers conflicts with results of Cheng *et al.*⁷ who report zero exchange bias for sputtered a -axis bilayers grown in a field at 200°C . It is likely that the facile chemical ordering we observe at room temperature is related to the use of a Ag seed film. Hsu *et al.*¹⁵ have demonstrated that chemical ordering of FePt (an L1₀ ordering alloy similar to MnPd) is enhanced by a Ag underlayer. We believe that the enhancement in chemical ordering is not due to a lattice strain effect since we find no evidence for interfacial lattice strain from either x-ray diffraction or electron microscopy studies. A possible alternative mechanism is that Ag reduces energy barriers to Mn and Pt surface diffusion during growth. These are known¹⁰ to control chemical ordering in epitaxial FePt.

It is interesting to compare the present results with those for a - and c -axis, MnPd/Fe bilayers, grown without a Ag seed layer on MgO(001) substrates by MBE⁶ and sputtering⁷ at much higher substrate temperatures. In the case of epitaxial bilayers grown by MBE, growth of MnPd at a substrate temperature of 200°C resulted in near complete chemical

ordering ($S \sim 1$) for both a and c -axis epitaxial samples grown simultaneously. Table I summarizes the properties (including interfacial coupling energy density, σ , estimated from $\sigma = H_E M_{\text{Fe}} t_{\text{Fe}}$ where M_{Fe} and t_{Fe} are the magnetization and thickness of the pinned Fe layer) of MBE-grown samples.

The spontaneously ordered MnPd/Fe bilayer of the present study has a lower interfacial coupling energy than the samples in which the MnPd was prepared at 200°C on Fe/MgO. This probably reflects the higher degree of chemical ordering. Secondly, the a -axis sample, grown on MgO, showed a larger interfacial coupling energy than the c -axis sample. Our x-ray diffraction measurements of peak widths (ΔQ) and rocking curve widths ($\Delta\Phi$) show that the structural coherence length for the c -axis sample is larger than for the a -axis sample. Using the integral breadth method, we estimate values of 12 nm and 10 nm, respectively. This implies a larger magnetic and structural domain size of the MnPd which may lead to a smaller interfacial pinning energy. This is supported by calculations of Li and Zhang.¹⁶ On the other hand, Cheng *et al.*⁷ report a -axis epitaxial samples show much smaller interfacial coupling energy than c -axis samples and suggest this may be due to a different spin structure of the MnPd interfacial plane. However, because of the high growth/annealing temperature (230°C – 300°C) in these latter experiments, it is unlikely the interfaces are atomically abrupt and an assumption of a bulk-like spin structure is probably not valid. The results of Cheng *et al.* may reflect a different degree of interdiffusion or a different length scale for the chemical ordering.

In summary, we have quantified spontaneous chemical ordering and interfacial pinning at room temperature in a -axis epitaxial MnPd(100) films on Fe(001)/Ag/Fe/GaAs(001). The identical magnetic structure of the Fe film at positive and negative field crossings contrasts with results¹⁴ for FeF₂/Fe bilayers, showing that different mechanisms of magnetic pinning operate for different materials.

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